

REMARKS

Claims 23, 54, 150-153, 242-244 and 247 have been amended, claims 245 and 246 have been canceled and new claims 248-300 have been added by this amendment. Upon entry of this amendment, claims 1-244 and 247-300 will be pending in the application.

\* Attached hereto is a marked-up version of the changes made by this amendment. The attached page is captioned "Version With Markings to Show Changes Made."

New claims 248-300 are essentially identical to claims 1-53 of U.S. Serial No. 09/408,296 that were finally rejected in the Office action mailed September 25, 2001. Pursuant to the suggestion by the Examiner in both of the above-mentioned Office actions, applicants have opted to continue prosecution of these claims in the subject application. In the remarks set forth below, applicants will address the rejections of claims 1-53 made in the above-mentioned Office action.

Rejections under 35 U.S.C. §102(b) - Nitrokemia

Reconsideration is respectfully requested of the rejection of claims 1, 3-5, 11-13, 15-22 and 200-211 and the subject matter of new claims 248, 250, 251, 256 and 258 (corresponding to claims 1, 3, 4, 9 and 11, respectively, of U.S. Serial No. 09/408,296) under 35 U.S.C. §102(b). The processes defined in the pending claims are submitted as novel over the disclosure in EP 0 019 445 (Nitrokemia).

The noble metal on carbon catalyst utilized in the oxidation processes defined in independent claims 1, 13, 248 and 256 is required to exhibit what applicants refer to as "deeply reduced" characteristics. Applicants have discovered that, by use of such deeply reduced catalysts that satisfy the various characterizing standards set forth in the claims, reagents can be effectively oxidized without undue loss of noble metal from the carbon support in the course of a catalytic oxidation reaction, particularly when conducted in an aqueous medium. The improved resistance to solubilization of the noble metal provided by using a deeply reduced catalyst in accordance with the present

invention overcomes a significant obstacle which has previously compromised the economic feasibility of using a noble metal on carbon support to catalyze oxidation reactions. Use of a deeply reduced noble metal on carbon catalyst in the oxidation of N-(phosphonomethyl)iminodiacetic acid (hereinafter "PMIDA") or a salt thereof provides for substantially quantitative oxidation to glyphosate or its salts and effective oxidation of the C<sub>1</sub> by-products of the reaction (formaldehyde and formic acid) to be maintained for a prolonged period and/or over numerous oxidation cycles without the excessive loss of noble metal as is otherwise typically encountered in acidic aqueous oxidation media.

The noble metal on carbon catalyst used to catalyze the oxidation reaction in independent claims 1 and 248 is characterized as yielding no more than about 1.2 mmole of carbon monoxide per gram of catalyst when a dry sample of the catalyst, after being heated at a temperature of about 500°C for about 1 hour in a hydrogen atmosphere and before being exposed to an oxidant following the heating in the hydrogen atmosphere, is heated in a helium atmosphere from about 20 to about 900°C at a rate of about 10°C per minute, and then at about 900°C for about 30 minutes. In alternative characterization, the deeply reduced catalyst employed in the processes defined in independent claims 13 and 256 comprises a carbon support having a noble metal, carbon and oxygen at a surface of the carbon support and has a ratio of carbon atoms to oxygen atoms of at least about 20:1 at the surface as measured by x-ray photoelectron spectroscopy after the catalyst is heated at a temperature of about 500°C for about 1 hour in a hydrogen atmosphere and before the catalyst is exposed to an oxidant following the heating in the hydrogen atmosphere.

Applicants confirm the observation by the Examiner in the Office action that the specific manipulations of the catalyst set forth in independent claims 1 and 13 with respect to how the carbon monoxide desorption yield and the surface C/O atomic ratio are to be determined are not process steps that have to occur before or during the oxidation of PMIDA to glyphosate or its

salts. Nonetheless, these manipulations define the deeply reduced characteristic of the catalyst utilized in the processes which, in turn, is an affirmative limitation of the claimed invention. Where the character of the catalyst is such that it meets the defined CO desorption or C/O surface ratio criteria, after the specified hour of heating in H<sub>2</sub>, the catalyst functions effectively without undue loss of noble metal, regardless of whether it is heated in H<sub>2</sub> before use in the claimed process.<sup>1</sup>

To provide a catalyst which gives such a low carbon monoxide desorption yield and/or exhibits such a high C/O atomic ratio at the surface of the carbon support, the catalyst is "deeply reduced" utilizing the catalyst preparation techniques described in the specification and particularly the working examples of the application. Applicants teach a variety of stratagems that may be employed to obtain a deeply reduced catalyst as claimed, including deoxygenating the carbon support prior to noble metal deposition, depositing the noble metal in one of its more reduced oxidation states (e.g., elemental platinum or a salt of Pt(II) instead of a salt of Pt(IV)) and reducing the surface of the catalyst after noble metal deposition by heating the surface (e.g., most preferably at temperatures of from about 550 to about 900°C) in a non-oxidizing environment (e.g., nitrogen, argon and helium) and/or exposing the surface to a reducing environment (e.g., formaldehyde, formic acid, amines, ammonium ions, and sodium borohydride (NaBH<sub>4</sub>)) or a gas-phase reducing agent (e.g., hydrogen, ammonia and carbon monoxide).

Nitrokemia discloses and claims a process for preparing glyphosate by batch oxidation of an aqueous mixture of PMIDA substrate with oxygen in the presence of a noble metal on carbon

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<sup>1</sup> It may be noted that the period of heating to achieve deep reduction is typically much longer than one hour; and that the combined time/temperature conditions normally used by Applicants to achieve deep reduction are more severe than the one hour at 500°C used to remove surface oxygen (presumably from oxidized promoter metals - see p. 21, line 24 to p. 22, line 7) pursuant to the test protocols for C/O desorption and C/O surface ratio. Cf. the working examples of the application.

catalyst. The charge mixture initially contains particulate PMIDA suspended in an apparently saturated aqueous solution thereof, the total initial concentration of PMIDA in the suspension being at least 7 g PMIDA per 100 ml water.

As acknowledged by the Examiner, Nitrokemia fails to disclose the carbon monoxide desorption yield and/or the surface C/O atomic ratio of the noble metal on carbon catalyst. Indeed, Nitrokemia contains no suggestion whatsoever that the catalyst should be subjected to reductive treatment to eliminate surface oxides, and in fact fails to even recognize the noble metal dissolution problem that would render the disclosed process commercially infeasible if practiced with a conventional noble metal on carbon catalyst. Accordingly, the processes defined in claims 1, 3-5, 11-13, 15-22, 200-211, 248, 250, 251, 256 and 258 cannot be said to be anticipated by the disclosure in Nitrokemia.

The Examiner's attempted reliance on the decisions in In re Best, 195 USPQ 430 (CCPA 1977) and In re Fitzgerald, 205 USPQ 594 (CCPA 1980) to overcome these deficiencies in the teaching of Nitrokemia is misplaced. The cited authority addresses the propriety of rejections under the narrowly applied doctrine of inherency. More particularly, these decisions stand for the proposition that

[W]here the Patent Office **has reason to believe** that a functional limitation asserted to be critical for establishing novelty in the claimed subject matter may, in fact, be an inherent characteristic of the prior art, it possesses the authority to require the applicant to prove that the subject matter shown to be in the prior art does not possess the characteristic relied on.

In re Fitzgerald, 205 USPQ at 597 (citing In re Best, 195 USPQ at 433) (emphasis added). In each of these cases, there was compelling evidence on the record that the features not specifically disclosed in the reference were in fact inherent, so that it made sense to require the applicant to prove otherwise. No such inference can reasonably be drawn from the disclosure of Nitrokemia, which fails to disclose any relevant information on

the method of preparation of any noble metal catalyst, much less any basis for the PTO to reasonably infer that the catalyst was "deeply reduced" in accordance with the instantly claimed criteria.

The In re Best decision illustrates the difference. There, the applicants appealed the Board's affirmance of a rejection of claims directed to a crystalline zeolitic molecular sieve catalyst composition useful in hydrocarbon conversion and to a process for producing them. The claim directed to the crystalline zeolite included two compositional requirements that were disclosed by the cited reference (Hansford) and four other parameters (cubic unit cell size, ion exchange capacity, oxygen adsorption capacity and X-ray powder diffraction pattern) that were not specifically disclosed by Hansford. The applicants urged that these latter four requirements were the unique result of their claimed process. However, with the exception of a functionally expressed rate of cooling,<sup>2</sup> all of the positive process limitations of the claimed process were expressly disclosed by Hansford. Because Hansford's calcined zeolite would necessarily have to be cooled to facilitate subsequent handling and the application suggested no criticality to the cooling rate, the Court found the Patent Office's assertion of inherency reasonable under these circumstances. In re Best 195 USPQ at 432-433. Where the claimed and prior art products are produced by identical or substantially identical processes, the court held that the PTO could require the applicant to rebut a finding of inherency (e.g., show that the cool-down rate for a typical laboratory-scale sample of zeolite prepared in accordance with Hansford's process using typical laboratory equipment would not

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<sup>2</sup> The process claim required "cooling the steamed zeolite to a temperature below 350°C at a rate sufficiently rapid that the cooled zeolite exhibits an X-ray powder diffraction pattern having the d-spacing corresponding to the Miller Indices, hkl, of 331 at least as strong in intensity as that corresponding to the Miller Indices 533, prior to any post-steaming ion exchange treatment."

necessarily or inherently yield a cooled zeolite with the requisite X-ray diffraction pattern or other claimed parameters). Id. at 433. In the absence of such a showing by the applicants, the Court affirmed the inherency rejection of both the composition and process claims.

By contrast, in the present situation, the Patent Office has failed to articulate any basis why it believes that the noble metal on carbon catalyst of Nitrokemia inherently possesses the carbon monoxide desorption yield, the surface C/O atomic ratio or other deeply reduced characteristics recited in the pending claims. As noted above, Nitrokemia is silent as to the problem of noble metal dissolution and **fails to teach or suggest any catalyst preparation methods, much less methods that include reductive treatment.** Accordingly, there is no reason to believe that the noble metal on carbon catalyst used in Nitrokemia satisfies the deeply reduced characteristics as claimed and Applicants should not be required to show otherwise.

Given the paucity of details in Nitrokemia regarding the noble metal on carbon catalysts<sup>3</sup> and the absence of any teaching as to how they are prepared, Applicants respectfully submit that, as a practical matter, a determination of whether the catalysts used in Nitrokemia possess the carbon monoxide desorption yield, the surface C/O atomic ratio or other deeply reduced characteristics is not possible. Numerous factors may influence whether a catalyst possesses the claimed deeply reduced characteristics, including the type of carbon support and how it was produced, handling and treatment of the carbon support before metal deposition, the source of the metals deposited on the carbon support, the metal deposition technique and chemistry, the components of the environment(s) to which the carbon support and catalyst precursor materials are exposed, agitation of the deposition solution, the temperature and duration of any heat

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<sup>3</sup> The only noble metal on carbon catalyst identified in Nitrokemia is a platinum or palladium on charcoal catalyst referred to as CARBO C Extra used in Examples 5-7.

treatment after metal deposition, the components of the atmosphere in which the catalyst is heated, the space velocity and gas pressure during heat treatment and how and in what environment the catalyst is cooled following heating steps among others. These factors are simply not disclosed at all in Nitrokemia. In the absence of such disclosure, a determination of whether the noble metal on carbon catalyst in Nitrokemia satisfy the deeply reduced characteristics called for in independent claims 1, 13, 248 and 256 is not possible by Applicants (or anyone else for that matter) and underscores the inappropriateness of the inherency rejection.

Accordingly, Applicants respectfully request that the rejection of claims 1, 3-5, 11-13, 15-22, 200-211, 248, 250, 251, 256 and 258 for lack of novelty be withdrawn.

**Rejections under 35 U.S.C. §103(a) - Nitrokemia**

Reconsideration is respectfully requested of the rejection of claims 1-22, 100-105 and 200-211 and the subject matter of new claims 248-251 and 256-258 (corresponding to claims 1-4 and 9-11, respectively, of U.S. Serial No. 09/408,296) under 35 U.S.C. §103(a). The processes defined in the pending claims are submitted as patentable over Nitrokemia.

Applicants note that, contrary to the observation at p. 2 of the Office action, not all of these claims are drawn to a continuous oxidation process; but a significant number of claims are so limited. Dependent claims 2, 14, 249 and 257 require that the oxidation process utilizing a deeply reduced noble metal on carbon catalyst be conducted in a continuous reactor system.<sup>4</sup> Independent claim 100 and dependent claims 101-105 are directed to a continuous process for the preparation of glyphosate or its salts by oxidation of PMIDA or a salt thereof. Although the

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<sup>4</sup> In Amendment B filed July 3, 2001, Applicants misstated that dependent claims 2, 14, 24, 34, 50, 54 and 80 require continuous oxidation of PMIDA to glyphosate in a continuous stirred tank reactor ("CSTR") (See Amendment B at page 33).

process of claim 100 is not limited to use of a deeply reduced catalyst, it requires continuously contacting a feed stream containing the PMIDA reagent with an oxygen source in a stirred tank reactor of a continuous reactor system (i.e., a CSTR) and in the presence of a particulate catalyst comprising a noble metal at the surface of a particulate carbon support.

As acknowledged by the Examiner, the disclosure in Nitrokemia is limited to a batch oxidation process and fails to teach conducting the oxidation of PMIDA in any continuous mode. In order to establish a *prima facie* case of obviousness, there must be some suggestion or motivation to modify the reference or to combine reference teachings. A teaching, suggestion or motivation to modify a reference or combine reference teachings is an essential element of an obviousness rejection. Furthermore, the modification or combination must not render the prior art teaching unsatisfactory for its intended purpose. Applicants respectfully submit that the Examiner has failed to establish a *prima facie* case of obviousness from Nitrokemia.

The only motivation to modify the teaching of Nitrokemia offered by the Examiner in the Office action is the vague statement that the artisan would be motivated to practice a continuous process for uninterrupted production of the product. No reference is cited in support of this contention. Although continuous processes do have known advantages, they also present complications which often more than offset the advantages. As a result, many reactions are preferably conducted in a batch mode, as is reflected by the prevalence of batch reactions in industrial chemical manufacturing facilities, and especially in the literature relating to the conversion of PMIDA to glyphosate.

With respect to claims 2, 14, 249 and 257, Nitrokemia offers no motivation either to conduct the reaction in a continuous manner or to use a deeply reduced catalyst of the type called for in each of these claims. These claims are submitted to be patentable on the basis of the use of a deeply reduced catalyst alone; but patentability is further supported by the requirement



of continuous oxidation in the presence of a heterogeneous catalyst.

Even assuming that the generalized desideratum of continuous processing might provide motivation to develop a continuous process for the oxidation of PMIDA to glyphosate in the presence of a heterogeneous catalyst, one skilled in the art would find Nitrokemia a peculiarly poor selection as a reference with which to start. Realistically, any extraneous impulse to develop a continuous process would be nullified by such selection, since development of continuous reaction would be wholly incompatible with the disclosure of this reference.

Nitrokemia describes a batch reactor having an initial charge comprising a slurry of PMIDA in a saturated solution thereof, which provides a very high initial concentration of PMIDA. This system is taught to be especially important to attainment of the higher specific conversions of PMIDA to glyphosate that the reference process purportedly realizes. Operation of a batch process in such manner allows substantial conversion to be achieved while the aqueous phase remains saturated with PMIDA. It is respectfully submitted that one skilled in the art would construe Nitrokemia as having achieved high productivity not only from the high payload resulting from the slurry charge, but also from the maximum PMIDA concentration driving force that is maintained over a substantial fraction of the batch cycle; and that the latter advantage would be uniquely associated with the batch mode in which the reaction is conducted. In fact, Nitrokemia seems even to suggest that the instantaneous reaction rate in a PMIDA slurry is somehow accelerated beyond that achieved in a saturated solution, though one skilled in the art might have reason to question a suggestion of that nature.

In any case, one skilled in the art would see no relevance of Nitrokemia to claims 100-105 which are directed to a process in which the oxidation of PMIDA is conducted in a continuous stirred tank reactor system in the presence of a particulate noble metal on carbon catalyst. In a continuous stirred tank

reactor, the reaction mixture is substantially back-mixed so that the reaction is conducted under terminal conditions, i.e., the concentration of PMIDA substrate remains constant at a level reflecting the conversion achieved in the reactor. To achieve substantial conversion of PMIDA substrate in such a system, the constant concentration of the PMIDA substrate is necessarily low relative to the product glyphosate, and for complete conversion the concentration of PMIDA approaches extinction. In fact, Nitrokemia itself teaches that the reaction is carried out essentially to the extinction of PMIDA, as is indeed necessary to achieve desired yield and facilitate recovery of glyphosate from the reaction mixture. Accordingly, the constant PMIDA concentration is far below saturation, even if a slurry is fed to the reactor. It is, therefore, unfathomable how one skilled in the art might apply the concept of Nitrokemia to a stirred tank reactor system. On the contrary, Nitrokemia would direct one skilled in the art away from a continuous stirred tank reactor, the employment of which would preclude adaptation of key features and principles on which the Nitrokemia process is based.

Accordingly, it is respectfully submitted that continuous process claims 2, 14, 100-105, 249 and 257 distinguish patentably over Nitrokemia under 35 U.S.C. §103(a). Moreover, with respect to the rejected claims which call for the use of a deeply reduced catalyst (claims 1-22, 200-211, 248-251 and 256-258), the *prima facie* case of obviousness is further lacking for the reasons set forth above in connection with the rejection under 35 U.S.C. §102(b).

In light of the fact that the Examiner's reliance on MPEP 2144.04, section V-E regarding the obviousness of making a batch process continuous is not expressly repeated in the present Office action, it is Applicants' understanding that such reliance has been withdrawn.

**U.S. Patent No. 3,969,398 (Hershman)**

In the Office action mailed January 11, 2001, then pending claims 1-199 were rejected under 35 U.S.C. §103(a) as

unpatentable over U.S. Patent No. 4,147,719 (Franz) in view of U.S. Patent No. 3,340,097 (Hess), U.S. Patent No. 3,969,398 (Hershman), Coloma et al. J. Catal. (1995) and U.S. Patent No. 5,627,125 (Ebner et al.).

In the response to this rejection (Amendment B, filed on July 3, 2001), Applicants alluded to disclosure in Monsanto patent references issued prior to Franz describing continuous processes for oxidation of PMIDA to glyphosate in a flow reactor comprising a fixed bed of carbon (See page 31 of Amendment B). Such disclosure is in fact found in Hershman. More specifically, Hershman suggests that the contacting of the aqueous solution of PMIDA with molecular oxygen and the activated carbon catalyst can be accomplished in a tubular continuous reactor packed with activated carbon (See col. 2, lines 20-21). In Example 5, the oxidation reaction was conducted in a lab scale continuous tubular reactor packed with activated carbon. Oxygen and a 4% by weight aqueous solution of PMIDA were introduced at the bottom of the reactor and flowed upwardly in concurrent fashion. The pressure and liquid flow rate were varied from 3.16 to 5.98 kg/cm<sup>2</sup> and from 3 to 15 cm<sup>3</sup>/min., respectively. Samples of the liquid reaction mixture withdrawn from the top of the reactor were dehydrated under reduced pressure to yield a solid product. Conversions were incomplete. Analysis of the solid product by gas liquid chromatography showed PMIDA content ranging from 6.5 to 65.3 weight percent and glyphosate content ranging from 21.0 to 82.1 weight percent (See col. 9, lines 15-33 and Table V).

Later in the Remarks of Amendment B, the continuous processes of claims 2, 14, 24, 34, 50, 54, 80 and 100 to 199 were distinguished from the disclosure in Hershman by the Applicants' observation that Hershman describes batch reactions (See Amendment B at page 34). Accordingly, given this inadvertent mischaracterization of Hershman, Applicants wanted to draw the Examiner's attention to the above-noted disclosure in this reference.

In any event, it is respectfully submitted that the combination of Hershman with Franz and/or the other references of

record does not render obvious a process for the preparation of glyphosate or a salt thereof by oxidation of PMIDA or a salt thereof using a deeply reduced noble metal on carbon catalyst as defined in independent claims 1, 13, 23, 33, 49 and 79, or a noble metal on carbon catalyst that further comprises a promoter as called for in independent claim 53. Accordingly, the patentability of dependent claims 2, 14, 24, 34, 50, 54 and 80 is not prejudiced by the continuous oxidation process disclosed in Hershman, nor by the previous mischaracterization of this reference inasmuch as the prior art disclosure of continuous conversion of PMIDA to glyphosate in a tubular fixed bed reactor was acknowledged in Amendment B.

The continuous process defined in independent claim 100 requires that the oxidation of PMIDA or a salt thereof be conducted in a stirred tank reactor of a continuous reactor system in the presence of a particulate catalyst comprising a noble metal at the surface of a carbon support and that the process further comprise separating the particulate catalyst from the product mixture by filtration. The disclosure in Hershman regarding the possibility of conducting the oxidation reaction in a continuous mode is limited to the use of a tubular reactor containing a fixed bed of activated carbon and in no way teaches or suggests conducting the oxidation reaction in a continuous reactor system comprising a CSTR in which PMIDA or salt thereof is continuously contacted with an oxygen source in the presence of a particulate catalyst that is subsequently filtered from the resulting product mixture. One skilled in the art would expect the Hershman fixed bed reactor to operate at least partially in plug flow, thereby providing a favorable kinetic driving force for the oxidation both of PMIDA and  $C_1$  by-products, formaldehyde and formic acid. The teachings of Hershman, therefore, would not render obvious the effectiveness of a CSTR system, each reactor of which operates under terminal conditions with respect to the conversion achieved therein. Accordingly, the continuous oxidation process defined in independent claim 100 and the claims

depending therefrom is patentably distinguished from the continuous oxidation process taught by Hershman.

**Supplemental Information Disclosure Statements**

Applicants filed a Fourth Supplemental Information Disclosure Statement on September 19, 2001. However, a copy of the PTO/SB/08A initialed by the Examiner did not accompany the Office action. Applicants respectfully request that an initialed copy accompany the next communication issued by the Examiner in connection with this application.

\* A Fifth Supplemental Information Disclosure Statement is enclosed submitting copies of the electrocatalyst references listed above that were not previously of record in this application and of International Publication No. WO 96/19485. WO 96/19485 discloses a process for preparing glyphosate by "wet oxidation" of PMIDA with an oxygen-containing gas in the absence of a catalyst. The process is preferably continuous and may be conducted in a pipe reactor (See Examples 5 and 6 and Fig. 2). It is respectfully submitted that this reference does not constitute an obstacle to the patentability of the processes defined in the pending claims.

**Patentability Over Nitrochemia is Supported by Consideration of Hershman and WO 96/19485**

Of references of record and known to Applicants' undersigned attorney, Hershman US 3,969,398 and WO 96/19485 contain the only disclosure of any continuous processes for the conversion of PMIDA to glyphosate. The Hershman disclosure appears in other Monsanto patents in essentially the same form. These references clearly fail to suggest any attempt to convert the Nitrochemia process to a continuous mode. Hershman would, if anything, deter any such notion.

It should be emphasized that neither Hershman nor WO 96/19485 describes a process in which PMIDA is oxidized in the presence of a noble metal on carbon catalyst. Hershman uses a carbon catalyst only. The PCT application uses no catalyst but

describes non-catalytic "wet oxidation" of PMIDA in a pipeline reactor.

Even by hindsight, one skilled in the art would be unlikely to attempt combination of the heterogeneous catalysis process of Nitrokemia with the non-catalytic process of the PCT application. But even if the combination were attempted, it would not yield the continuous stirred tank reactor system of claims 100-199.

As to Hershman, one skilled in the art would anticipate unsatisfactory complications, e.g. plugging of the reactor, in any attempt to implement the PMIDA slurry process of Nitrokemia in a fixed bed reactor. In fact, under the conditions described in the Nitrokemia working examples, the reaction product would constitute a slurry of glyphosate, thereby compounding the problem of maintaining flow through a fixed bed reactor. Thus, the teachings of Hershman reinforce the conclusion that the process of Nitrokemia should be practiced in a batch mode only. Considered in light of the art actually describing continuous oxidation of PMIDA, Nitrokemia leads away from the instantly claimed invention, especially the continuous stirred tank reactor system of claims 100-199.

**Rejections under 35 U.S.C. §103(a) - Nitrokemia, Jalan and Itoh**

Reconsideration is respectfully requested of the rejection of claims 1-247 and the subject matter of new claims 248-300 (corresponding to claims 1-53 of U.S. Serial No. 09/408,296) under 35 U.S.C. §103(a). The processes defined in the pending claims are submitted as patentable over Nitrokemia in view of U.S. Patent Nos. 4,186,110 (Jalan et al.) and 5,876,867 (Itoh et al.).

Jalan '110 and Itoh '867 are among a group of several U.S. patents now of record in this application (listed below) that disclose electrocatalysts comprising a platinum alloy dispersed on a carbon powder support and methods for their preparation. The electrocatalyst powder may be bound to a suitable supporting member or substrate to form an electrode said to be useful in fuel cell or used in other applications. One or more metals are

alloyed with the platinum and may include aluminum, titanium, cerium, iron, cobalt, nickel and manganese among others. The process by which the supported platinum alloy is obtained comprises depositing a suitable platinum compound and a compound(s) of the alloying metal(s) onto the carbon powder carrier and subsequently subjecting the catalyst precursor to a reductive alloying treatment by heating it to a temperature of 600°C or higher. Some of these references disclose an alloying step in which the catalyst precursor is heated in a reducing atmosphere such as hydrogen.

4,186,110 (Jalan, et al.)  
4,970,128 (Itoh, et al.)  
5,024,905 (Itoh, et al.)  
5,079,107 (Jalan)  
5,096,866 (Itoh, et al.)  
5,178,971 (Itoh, et al.)  
5,189,005 (Watanabe, et al.)  
5,225,391 (Stonehart, et al.)  
5,759,944 (Buchanan, et al.)  
5,876,867 (Itoh, et al.)

With respect to the pending claims which require the use of a deeply reduced catalyst characterized by carbon monoxide desorption yield (claims 1-12, 75-78, 150-199, 200-205 and 248-255) or the surface C/O atomic ratio, either before or after noble metal deposition (claims 13-32, 38-52, 66-74, 79-99, 155-199, 206-217, 224-229, 236-241, 256-269, 284 and 288-300), Applicants respectfully submit that these limitations are not disclosed in the electrocatalyst references either expressly or inherently. The present application is a divisional of U.S. Serial No. 09/248,655 which contains claims directed to the deeply reduced catalysts and processes for their preparation. A full discussion of the relevance of these references to the deeply reduced characteristics of the catalyst used in the

practice of the present invention is contained in Amendment E, filed December 19, 2001 in connection with U.S. Serial No.

\* 09/248,655 (copy enclosed).

Applicants acknowledge that forming an oxidation catalyst by a process comprising depositing a noble metal at a surface of a carbon support and subsequently heating the surface at a temperature of at least about 400°C (as required in independent claims 33 and 270) or exposing the surface to a reducing environment (as required in independent claims 49 and 288) are arguably taught in one or more of the above-mentioned electrocatalyst references.

**Further Distinction of Claimed Process over Nitrokemia in view of Itoh and Jalan**

Applicants further respectfully submit that there is no sound basis for combining Jalan, the Itoh patents, Watanabe, Stonehart, Buchanan, or any combination thereof with Nitrokemia; and further that, even if by hindsight such combination were made, it would still fail to yield the patented invention.

**Oxidation with Deeply Reduced Catalyst vs. Nitrokemia, Jalan, Itoh, etc.**

None of the cited references contain any teaching or suggestion of the CO desorption characteristics of the catalyst used in the processes of claim 1 or 248, or the C/O surface ratio of the catalyst used in the processes of claim 13 or 256. Nor do the references contain any teaching of the oxidation of a substrate using a catalyst of the type prepared in the manner defined in claims 49 or 262. As noted, attached Amendment E as submitted in copending Ser. No. 09/248,655 discusses the question of whether the catalysts as prepared by Jalan or Itoh may inherently possess the characteristics of catalysts as defined in certain of these claims. It is respectfully submitted that inherency cannot be shown.

However, assuming *arguendo* that a catalyst of Jalan or Itoh inherently possessed the properties of one or more of claims 1, 13, 49, 248, 256 or 262, it is respectfully submitted that no



basis is provided for obviousness of the instantly claimed processes in which a substrate is oxidized by contacting it with the catalyst and oxygen. Jalan and Itoh relate to fuel cell catalysts, particularly for use in cathodes for the reduction of oxygen, though the Itoh reference may in one instance suggest use of the catalyst in an anode for oxidation of a fuel. Where used as a cathode, the catalyst serves to facilitate the reduction of oxygen, but does not function by contact with the substrate (fuel). Where used as an anode, the catalyst serves to facilitate the oxidation of the fuel, but does not function by contacting the fuel in the presence of oxygen. In any case, the references offer little by way of description of the fuel cells, and particularly fail to describe any cell in which contact of catalyst with both oxygen and substrate might even incidentally occur.

Applicants recognize that, while Itoh and Jalan are primarily concerned with fuel cells, Jalan makes passing mention of the possibility of using the catalysts therein described in "chemical, pharmaceutical, automotive and anti-pollution fields." However, there is no teaching in any of these references of a chemical oxidation reaction in a system which can "solubilize a noble metal," as called for in claims 248-300, and certainly no remote suggestion of the use of such catalyst in the oxidation of PMIDA to glyphosate, as required by the remaining claims of the application. This is significant because noble metal on carbon catalysts are well known for use in other types of reactions, prominently hydrogenations, but are not commonly used in liquid phase oxidations, largely for the reasons explained above, i.e., the tendency of noble metal to be leached into an aqueous medium, especially an acidic aqueous medium. Jalan's reference to "automotive and anti-pollution fields" is unelaborated, but may well relate to gas phase reactions rather than to reactions in a system which can solubilize a noble metal.

It is further respectfully noted that, even if Jalan and Itoh were construed as describing catalysts which inherently meet the claimed criteria of a deep reduction, the putative inherency

is not manifest but, at most, latent. Since the claimed oxidation reactions are not described, there is no basis for rejection under §102; and since the deep reduction characteristics are not described, there can be no basis for rejection under §103. Obviousness cannot be predicated on what is unknown, *in re Spormann*, 150 U.S.P.Q. 449, 452 (C.C.P.A. 1966).

Jalan and Itoh particularly lack any relevance to claims calling for the oxidation of PMIDA to glyphosate. It would plainly require a most liberal application of hindsight to reconstruct those claims from the combination of Jalan, Itoh and Nitrokemia based on putative latent inherency in the properties of the Jalan or Itoh catalyst.

Claims to Continuous Process vs. Nitrokemia, Itoh, Jalan

Jalan and Itoh add nothing to the teachings of Nitrokemia with regard to the claims which call for continuous oxidation of a substrate. Thus, even if the references are combined, they fail to teach a continuous process of any nature.

As noted above, one skilled in the art would be deterred from conducting Nitrokemia's heterogeneous catalytic oxidation in a continuous fashion. Because it emphasizes the importance of conducting the reaction in a PMIDA slurry, Nitrokemia teaches away from both the continuous stirred tank reactor system of claim 100, which necessarily operates under terminal conditions with low PMIDA concentration (even if fed with a PMIDA slurry), and for that matter from the tubular fixed bed reactor of Hershman which might be expected to suffer serious plugging problems if fed with a slurry.

The deficiencies of Nitrokemia are not met by Jalan or Itoh. Neither Jalan nor Itoh remotely deals with the problems that would confront one skilled in the art in approaching the development of a continuous process for the manufacture of glyphosate from PMIDA. These references utterly fail to offer any way to implement the slurry process of Nitrokemia in any catalytic reaction system.

### Promoted Catalysts

Claims 53-99 and 144-199 call of the use of a noble metal on carbon catalyst that further comprises a promoter. All of these claims are limited to the preparation of glyphosate from PMIDA. Promoters have been found to significantly enhance the effectiveness of the catalyst for oxidation of substrates such as PMIDA, and/or by-products such as formaldehyde and formic acid, in an acidic aqueous medium.

The use of a promoter further enhances the effectiveness of the continuous process for the oxidation of PMIDA to glyphosate, as defined in claims 54 and 144-199.

Applicants acknowledge that some of the metals alloyed with platinum in the electrocatalyst references may serve as promoters in the practice of the claimed invention.

However, as discussed above, neither Jalan nor Itoh offers any suggestions relevant to oxidation of PMIDA to glyphosate. Jalan alludes to the use of promoters for other reactions, but offers no detail; Itoh offers even less than Jalan. Moreover, the principal function of the alloying metal in Itoh is in a process for producing a skeletal platinum catalyst by leaching out the alloying metal. In any event, it is respectfully submitted that no combination of Nitrokemia, Itoh and/or Jalan establishes obviousness of the process of claims 53-59 or 144-199.

### Further Comment re Rejections over Nitrokemia, Itoh and Jalan

Applicants note that various dependent claims include significant features that further distinguish the claimed processes from the references of record. The Office action dismisses the significance of these features without any citation of prior art.

It is respectfully submitted that the patentability of the independent claims pending herein is amply established by the considerations that are discussed hereinabove, thereby mooting the grounds on which the various dependent claims stand rejected. Applicants, therefore, do not propose to extend the record by

detailed discussion of additional dependent claims at this time. However, Applicants respectfully traverse the finding of unpatentability of the dependent claims and reserve the right to further prosecute these claims in further proceedings herein.

Drawings

In response to the objections raised in the Notice of Draftperson's Patent Drawing Review attached to the Office action, a Letter to the Official Draftsman submitting formal drawings is enclosed.

\*

CONCLUSION

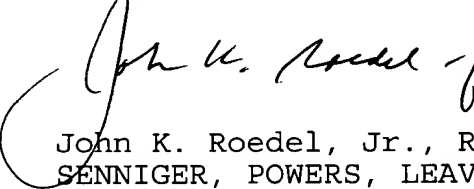
In view of the above, it is respectfully submitted that the pending claims are clearly patentable over the art of record.

Favorable reconsideration and early allowance of all claims are respectfully solicited.

A check is enclosed to the cover the \$1,590.00 additional claim fee. The Commissioner is authorized to charge any fee deficiency in connection with this amendment or refund any overpayment to Deposit Account No. 19-1345.

\*

Respectfully submitted,



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VMK/JKR/msc/cwa  
\*Attachments

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VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE CLAIMS

**Claim 23 has been amended as follows:**

23. (twice amended) A process for the preparation of N-(phosphonomethyl)glycine or a salt thereof, the process comprising contacting N-(phosphonomethyl)iminodiacetic acid or a salt thereof with an oxidation catalyst in the presence of oxygen, wherein the catalyst comprises a carbon support comprising: (a) a noble metal at a surface of the carbon support; and (b) a surface layer having a thickness of about 50 Å as measured inwardly from the surface and comprising carbon and oxygen, the ratio of carbon atoms to oxygen atoms in the surface layer being at least about 20:1 as measured by x-ray photoelectron spectroscopy after the catalyst is heated at a temperature of about 500°C for about 1 hour in a hydrogen atmosphere and before the catalyst is exposed to an oxidant following the heating in the hydrogen atmosphere.

**Claim 54 has been amended as follows:**

54. (once amended) The process of claim 53 wherein the process is [**contacted**] conducted in a continuous reactor system.

**Claim 150 has been amended as follows:**

150. (twice amended) A process as set forth in claim 147 wherein, before said oxidation of said reagent, the catalyst is characterized such that no more than about 1.2 mmole of carbon monoxide per gram of catalyst is desorbed when a dry sample of the catalyst, after being heated at a temperature of about 500°C for about 1 hour in a hydrogen atmosphere and before being exposed to an oxidant [**after**] following the heating in a hydrogen atmosphere, is heated in a helium atmosphere from about 20 to about 900°C at a rate of about 10°C per minute, and then at about 900°C for about 30 minutes.

**Claim 151 has been amended as follows:**

151. (twice amended) A process as set forth in claim 150 wherein, before said oxidation of said reagent, the catalyst is characterized such that no more than about 0.7 mmole of carbon monoxide per gram of catalyst is desorbed when a dry sample of the catalyst, after being heated at a temperature of about 500°C for about 1 hour in a hydrogen atmosphere and before being exposed to an oxidant [after] following the heating in a hydrogen atmosphere, is heated in a helium atmosphere from about 20 to about 900°C at a rate of about 10°C per minute, and then at about 900°C for about 30 minutes.

**Claim 152 has been amended as follows:**

152. (twice amended) A process as set forth in claim 151 wherein, before said oxidation of said reagent, the catalyst is characterized such that no more than about 0.5 mmole of carbon monoxide per gram of catalyst is desorbed when a dry sample of the catalyst, after being heated at a temperature of about 500°C for about 1 hour in a hydrogen atmosphere and before being exposed to an oxidant [after] following the heating in a hydrogen atmosphere, is heated in a helium atmosphere from about 20 to about 900°C at a rate of about 10°C per minute, and then at about 900°C for about 30 minutes.

**Claim 153 has been amended as follows:**

153. (twice amended) A process as set forth in claim 152 wherein, before said oxidation of said reagent, the catalyst is characterized such that no more than about 0.3 mmoles of carbon monoxide per gram of catalyst is desorbed when a dry sample of the catalyst, after being heated at a temperature of about 500°C for about 1 hour in a hydrogen atmosphere and before being exposed to an oxidant [after] following the heating in a hydrogen atmosphere, is heated in a helium atmosphere from about 20 to about 900°C at a rate of about 10°C per minute, and then at about 900°C for about 30 minutes.

Claim 242 has been amended as follows:

242. (once amended) A process as set forth in claim 100  
[comprising:

contacting N-(phosphonomethyl)iminodiacetic acid or a salt thereof with said catalyst and oxygen, thereby producing a used catalyst and a liquid reaction product comprising N-(phosphonomethyl)glycine or a salt thereof;

separating said liquid reaction product from said used catalyst;  
and

contacting] wherein the used particulate catalyst separated from the product mixture is recycled to the continuous reactor system and additional N-(phosphonomethyl)iminodiacetic acid or a salt thereof is contacted with said used catalyst and oxygen, thereby producing additional [liquid reaction] product mixture comprising N-(phosphonomethyl)glycine or a salt thereof.

Claim 243 has been amended as follows:

243. (once amended) A process as set forth in claim 242 further comprising:

separating said [additional liquid reaction product from said] used particulate catalyst from said additional product mixture by filtration.

Claim 244 has been amended as follows:

244. (once amended) A process as set forth in claim 243 further comprising repetitively contacting said used particulate catalyst with oxygen and further additional N-(phosphonomethyl)glycine or a salt thereof to produce further additional [liquid reaction] product mixture.

Claim 247 has been amended as follows:

247. (once amended) A process as set forth in claim 242 wherein said [catalyst comprises a particulate noble metal on carbon] particulate catalyst [that] is slurried in a liquid reaction medium within said stirred tank reactor, said liquid reaction medium comprising N-(phosphonomethyl)iminodiacetic acid or a salt thereof[, said used catalyst being separated from said liquid reaction product by filtration].

Claims 245 and 246 have been cancelled.

New claims 248-300 have been added.